Polarity of GaN Grown on Sapphire by Molecular Beam Epitaxy with Different Buffer Layers

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We report on polarity of GaN films grown on sapphire substrates by molecular beam epitaxy using different buffer layers and growth conditions. On high temperature AlN or GaN buffer layers, the GaN films typically show Ga- or N-polarity, respectively. When low temperature (either AlN or GaN) buffer layers are employed, GaN films of both polarities can be grown, but these films have high density of inversion domains. Insertion of additional GaN/AlN quantum dot layers between the buffer layers and the GaN films provides strain relief and a significant improvement in the quality of the GaN epilayers.

Introduction Wurtzite GaN epilayers are predominantly grown on sapphire substrates by metalorganic chemical vapor deposition (MOCVD) or molecular beam epitaxy (MBE). In addition to the lattice and thermal mismatches, GaN does not share the same atomic stacking order with sapphire [1]. Consequently, depending on the growth conditions and the buffer layers used, the [0001] or c direction of the film can be either parallel or anti-parallel to the growth direction, resulting in GaN films of two different polarities, Ga- or N-polarity, respectively. Mixed polarity regions or inversion domains can also present in GaN films. It is observed that Ga- and N-polar films have vastly differing surface, structural, optical, and electrical properties [2]. Therefore, the proper control of film polarity during growth, and the characterization of films with different polarities, becomes very important in GaN-based device epitaxy.

Generally, a Ga-polar film is obtained when it is grown on low temperature (~600^{o}C) buffer layers (either AlN or GaN) on sapphire substrates by MOCVD. In the case of MBE, a high temperature (900^{o}C) AlN buffer layer may lead to Ga-polar GaN films [3]. Otherwise, the correlation between film polarity and MBE growth is not well understood. In this paper, we report on MBE-grown GaN films on sapphire substrates which have different polarities when grown on various buffer layers under different growth conditions.

Experiments GaN layers were grown by MBE on c-plane sapphire substrates using radio frequency activated N. Four groups of samples were grown and investigated. The first and second sets utilized GaN buffer layers grown at low (~500 ^{o}C) and high (~800 ^{o}C) temperature. The third and fourth groups utilized AlN buffer layers, again grown at low (~500 ^{o}C) and high (~930 ^{o}C) temperature. In addition to growth temperature, the growth rate of the buffer layer was also changed in this investigation. Following the buffer layers, typically 1 µm thick GaN layers were grown at substrate temperatures between 720 and 850 ^{o}C with growth rates in the range of 300 to 1000 nm per hour under N-limited (Ga-rich) conditions. Some additional GaN/AlN and quantum dot (QD) layers may be inserted between the buffer and the GaN film in order to relieve strain and improve the crystal quality.

The polarity of the GaN films was determined by wet chemical etching (in 160 ºC H_3PO_4) and the surface morphologies of both as-grown and etched surfaces measured by AFM [4]. Sample polarity determined from etching and AFM images was in agreement with the in-situ RHEED patterns observed during sample growth. A well-known method based on convergent beam electron diffraction (CBDE) studies was also applied to a few typical samples to confirm the polarity assignment. In addition, conventional transmission electron microscopy (TEM) studies were carried out in order to examine the details of the microstructure of the films. Both CBED and TEM studies were performed using the TOPCON 002B electron microscope operated at 200 keV.

Results To summarize the main results, we list in Table I some of the GaN epilayer samples investigated having various polarities and grown on different types of buffer layers. In addition to growth temperature, growth rate, and buffer layer thickness, the width (full width at half maximum or FWHM) of both symmetric [002] and asymmetric [104] x-ray diffraction (XRD) peaks are listed. The XRD spectra were measured with a Philips X'Pert MRD system equipped with a four-crystal Ge (220) monochromator.

TABLE I. Samples with different buffer layers and polarities. T_b, d_b and R_b are, respectively, the growth temperature, thickness and growth rate of buffer layers. $\Gamma_{[002]}$ and $\Gamma_{[104]}$ are the FWHM of the [002] and [104] XRD peak respectively.

Sample #	Buffer	T_b (^{o}C)	d_b (nm)	R_b (nm/hr)	$\Gamma_{[002]}$ (arcmin)	$\Gamma_{[104]}$ (arcmin)	Polarity
381	AlN	914	18	55	2.0	5.0	Ga
643	AlN	920	30	60	2.2	6.3	Ga
687	GaN	900-800	40	600	9.1	6.6	N
688	GaN	900-800	30	600	9.6	6.1	N
666	AlN	500	20	60	1.9	5.1	Ga
698	AlN	500	20	60	6.3	24.0	Ga
630	AlN	510	14	28	13.5	9.2	N
636	AlN	510	12	25	3.7	4.5	N
668	GaN	500	150	600	7.2	11.5	Ga
680	GaN	500	60	600	5.6	15.5	Ga
618	GaN	505	220	220	5.3	5.2	N
622	GaN	500	220	220	9.3	4.6	N

The results show that the GaN films grown on high temperature (~900 ºC) AlN buffer layers, with thicknesses in the range of 8-35 nm and growth rates of 50-60 nm/hour, have Ga-polarity (sample Nos. 381 and 643). Conversely, the GaN films grown on high temperature (~ 850 ^{o}C) GaN buffer layers, with thickness ~30-40 nm and growth rate of 600 nm/hour, turned out to be N-polarity (sample Nos. 687 and 688). For the GaN films grown on low temperature (~500 ºC) buffer layers (either AlN or GaN), the samples were found to have either Ga- or N-polarity, depending on the growth rate. When 20 nm thick AlN buffer layers grown at a rate of 60 nm/hour were employed, Ga-polarity films resulted (sample Nos. 666 and 698). However, when ~10-20 nm thick AlN buffer layers are grown at a lower rate of 20-30 nm/hour, N-polarity films can be produced (sample Nos. 630 and 636). For GaN films grown on low temperature (~500 ºC) GaN buffer layers, their polarities also depended upon the growth rate of the buffer layer. A 60-150 nm thick GaN buffer layer grown at a rate of ~ 600 nm/hour can lead to Ga-polarity (sample Nos. 668 and 680). When ~110-220 nm thick GaN buffer layers were grown at 500 ºC with a lower growth rate (220 nm/hour), the resultant layers were of N-polarity (sample Nos. 618 and 622).

CBED studies performed on sample 381 showed that it was almost completely of pure Ga-polarity, as shown in Figure 1. TEM studies showed that there were narrow (diameter of a few nm) inversion domains occasionally present in this layer. Their density was very low and estimated to be ~$1\text{x}10^{7}\text{cm}^{-2}$. CBED and conventional TEM studies were also carried out for sample No. 618 (see Figure 2) and an essentially N-polar film was demonstrated. However, the density of inversion domains in that sample was much higher and estimated to be ~$1\text{x}10^{11}\text{cm}^{-2}$. All of the inversion domains in both types of samples were along the growth direction and propagated from the substrate/buffer interface up to the epilayer surface. The diameter of the inversion domains was in the range of ~5-10 nm. Some pits associated with inversion domains were also observed in the Ga-polar sample. As compared to the Ga-polar sample No. 381, the observed dislocation density in the N-polar sample No. 618 is much lower (~$5\text{x}10^{9}\text{cm}^{-2}$ and ~$5\text{x}10^{8}\text{cm}^{-2}$ near the surface of sample Nos. 381 and 618, respectively).

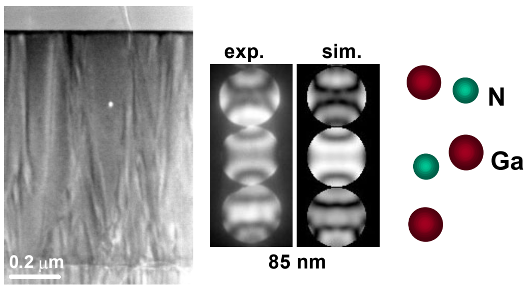

Fig. 1. Cross-section TEM image with experimental and simulated CBED patterns for sample 381

As presented in Table I, the width of the symmetric [002] XRD peak is more sensitive to the growth of the buffer layer than that of the asymmetric [104] peak. For the Ga-polar samples, the [002] peaks are always narrower than the [104] peak. Whereas for N-polar films, broader [002] peaks were commonly observed. In addition, the GaN films grown on AlN buffer layers typically have sharper [002] peaks as compared to those on GaN buffers. The narrowest [002] peak was observed from the sample grown on a high temperature AlN buffer.

In marked contrast to the generally held belief that N-polar films are of “poorer” quality, the observed PL efficiencies of N-polar samples are in fact higher than those of Ga-polar films. Generally, more than an order of magnitude higher PL efficiency was observed from typical N-polar films as compared to Ga-polar samples. This observation suggests that the high density of inversion domains observed in N-polar films may be not responsible for the nonradiative recombination of photo-carriers.

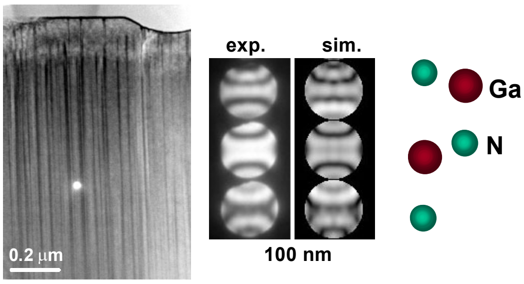

Fig. 2. Cross-section TEM image with experimental and simulated CBED patterns for sample 618

We also investigated the effects of buffer layer growth on the formation of defects in Ga-polar GaN films. For this purpose, additional GaN/AlN and quantum dots layers were inserted between the high temperature AlN buffer and the GaN films. With proper control of the growth conditions, a low density of etched pits (~$5x10^7cm^{-2}$) were revealed by the wet chemical etching and AFM images from the samples when additional GaN/AlN and QD layers were inserted. These results show that high quality GaN films with low dislocation density can be grown by MBE with the proper design and growth of buffer layers.

Conclusion We found that the GaN films grown on high temperature AlN and GaN buffer layers on sapphire substrates by MBE show Ga- and N-polarity, respectively. However, the polarity of GaN films grown on low temperature buffer layers depends critically upon the growth conditions. With increasing growth rate of the buffer layer, a GaN film having Ga-polarity is more likely to be grown.

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